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Title: Measurements of Extinct Fission Products in Nuclear Bomb Debris: Determination of the Yield of the Trinity Nuclear Test 70 Years Later

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Abstract: This contribution describes an approach to measuring extinct fission products that would allow for the characterization of a nuclear test at any time. The isotopic composition of molybdenum in five samples of glassy debris from the 1945 Trinity nuclear test has been measured. Non-natural molybdenum isotopic compositions were observed, reflecting an input from the decay of the short-lived fission products ^{95}Zr and ^{97}Zr. By measuring both the perturbation of the ^{95}Mo/^{96}Mo and ^{97}Mo/^{96}Mo isotopic ratios and the total amount of molybdenum in the Trinity nuclear debris samples, it is possible to calculate the original concentrations of the ^{95}Zr and ^{97}Zr isotopes formed in the nuclear detonation. Together with a determination of the amount of plutonium in the debris, these measurements of extinct fission products allow for new estimates of the efficiency and yield of the historic Trinity test.

Significance statement: This work describes an approach to post-detonation nuclear forensics involving isotopic measurements that allows for characterization of a nuclear detonation at any time. By performing high precision measurements of stable isotope perturbations in nuclear bomb debris, it is possible to quantify short-lived fission products long after they have decayed below radiometric detection limits and become extinct. The extinct fission product concentrations can be used to reconstruct details of the nuclear device months to years after the detonation occurred. The approach is demonstrated by analysis of debris from the Trinity nuclear test and new estimates of the efficiency and yield of the historic test are presented.

Text: The ability to confirm the occurrence and nature of a nuclear test is essential to modern nuclear treaty monitoring.(1) A robust multination monitoring system is in place for underground nuclear tests, relying primarily on seismic signals and other prompt measures.(1-3) Radiochemical measurements of actinides and fission products are also an integral part of treaty monitoring, but these too are time sensitive, relying on immediate access to samples containing short-lived radionuclides.(3) In particular, the most useful refractory peak-yield fission products are short-lived and no longer detectable radiometrically in the debris from older tests (when months or years have passed since the test). An ability to determine the concentration of extinct fission products in old debris would allow for independent characterization of a nuclear test at any time and could serve as a valuable additional tool for ongoing nuclear non-proliferation and

verification endeavors. We report here the first determination of the number of fissions that occurred in a nuclear test via high precision mass-spectrometric measurements of perturbation in ^{95}Mo and ^{97}Mo, which are the stable decay daughters of ^{95}Zr and ^{97}Zr fission products. These fission determinations are combined with plutonium measurements to estimate the efficiency and yield of the first nuclear test, Trinity.

Seventy years have passed since the Trinity test and the exact yield and performance of the first atomic bomb are still debated. Many estimates of the bomb yield have been published, ranging from 8-61 kT, with the official U.S. Department of Energy yield of 21 kT based on historic radiochemical measurements.(4-6) To determine the plutonium fission efficiency and yield of the Trinity device, a measurement of residual unreacted plutonium and the number of fissions was used (equation 1).(7) Although radiochemical measurements were performed in 1945, they were crude relative to modern standards, leaving some uncertainty in the official yield.(8)

$$Efficiency = \frac{F_{Total}}{Pu_{Ingoing}} = \frac{f_{Sample}}{f_{Sample} + Pu_{Sample}} \qquad \text{(Equation 1)}$$

F_{Total} = total number of fissions in the device

$Pu_{Ingoing}$ = total number of plutonium atoms in the device

f_{sample} = number of fissions measured in the debris sample

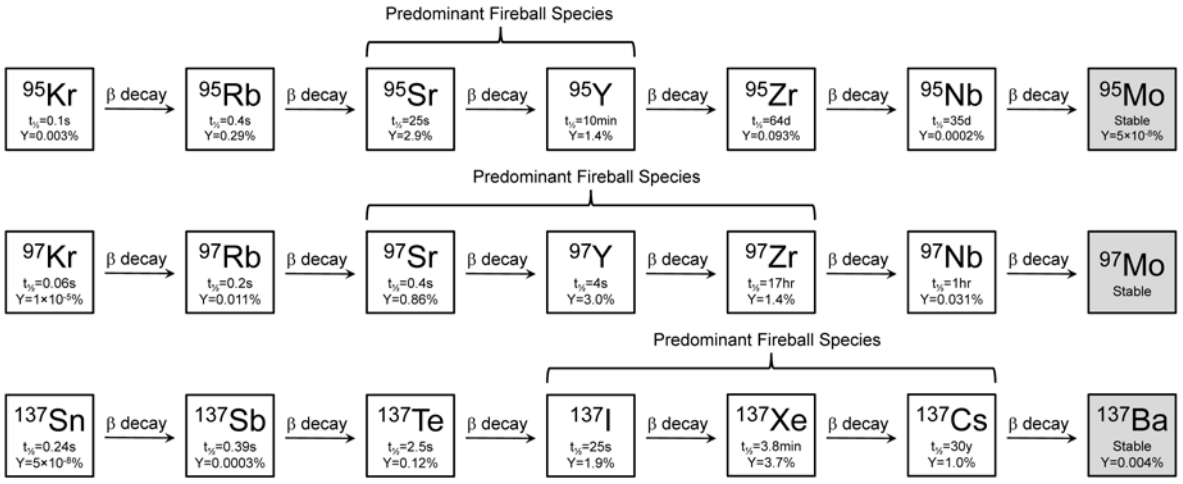
Pu_{sample} = number of plutonium atoms measured in the debris sample

While long-lived fission products such as ^{137}Cs and ^{90}Sr are detectable in Trinity nuclear debris today,(9-14) these radionuclides have volatile precursors (e.g. ^{137}I and ^{137}Xe; Figure 1a) and are known to fractionate severely from Pu, making reliable interpretation difficult.(5, 15) In contrast, refractory peak-yield fission products such as ^{95}Zr ($t_{1/2}$ = 64.02 days) and ^{97}Zr ($t_{1/2}$ = 16.75 hours) allow for more accurate determination of the bomb efficiency and yield, but have long since decayed below detection limits and are termed extinct radionuclides.

We hypothesized that it might be possible to reconstruct the original concentrations of ^{95}Zr and ^{97}Zr fission products by measuring the isotopic composition of the stable molybdenum in Trinity nuclear debris. Molybdenum has seven stable isotopes. Four of these isotopes, ^{95}Mo, ^{97}Mo, ^{98}Mo, and ^{100}Mo are produced by fission via a combination of independent fission yield

and β decay along each mass chain, as shown by the diagonal arrows in Figure 1b. In contrast, ^{92}Mo, ^{94}Mo, and ^{96}Mo are blocked from perturbation by fission product β decay, as each β decay chain terminates at a stable zirconium nuclide. There are no contributions to ^{92}Mo and ^{94}Mo from independent plutonium fission yield, and the independent fission yield of ^{96}Mo is less than 2.5×10^{-6} percent.(16) Therefore, the presence of fissions in a sample will be evidenced by a perturbation of specific molybdenum atom ratios. For example, Trinity nuclear debris should display an elevated ^{95}Mo/^{96}Mo atom ratio when compared to the known natural abundance. A similar concept has been used to demonstrate the presence of extinct fission products in the Oklo reactor of Southwest Africa.(17, 18) A challenge of our approach, however, is that only a small amount of Mo is formed by the nuclear fission process compared to the larger amount of natural Mo present in the rock and soil that is entrained in the fireball and mixed with the glassy debris.(19) In contrast to the landmark Oklo studies, the Trinity nuclear debris samples are many times more dilute in actinide and fission fragment concentration (~0.6 μg/g ($Pu_{trinitite}$) vs. ~0.5 g/g (U_{Oklo})), rendering any potential isotopic perturbation much smaller.(18)

A



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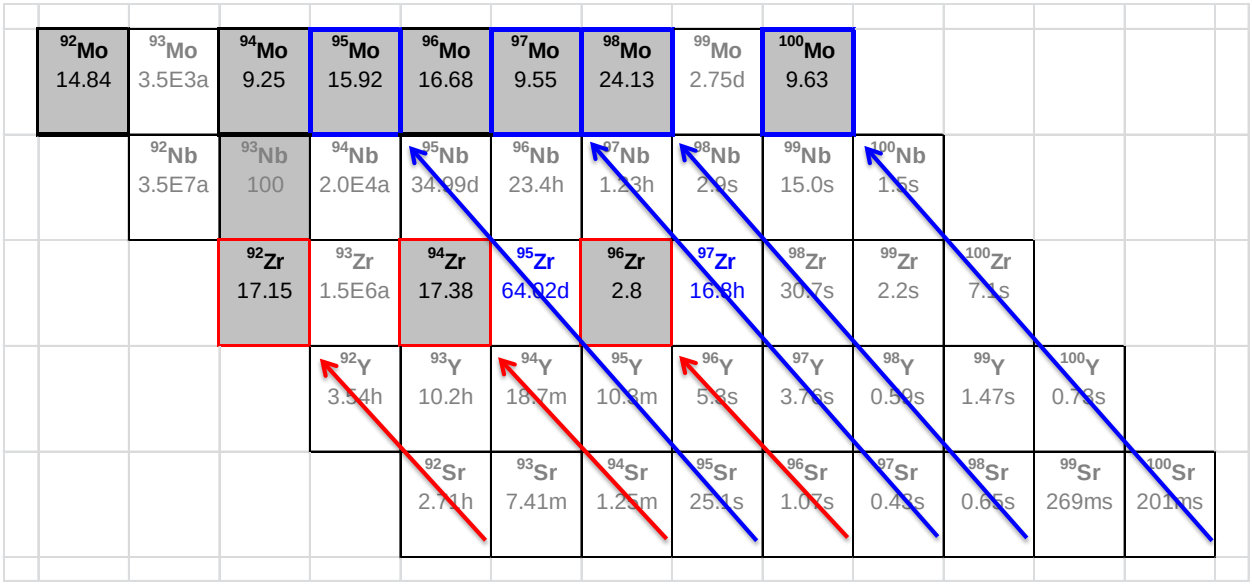

Figure 1. (A) Decay schemes for mass 95, 97, and 137 mass chains for plutonium fission. The half-life and independent chain yield for the fission spectrum in plutonium is shown. (B) Partial scheme for the decay of fission products into stable molybdenum and zirconium species. Stable species are denoted with gray boxes, and the natural isotopic abundance is shown for each stable species. Radioactive species and their half-lives are shown in white boxes. Blue and red arrows

illustrate β decay chains of interest, and the bolded blue and red boxes indicate stable isotopes that are perturbed by decay of short lived fission products.

Five samples of Trinity nuclear debris (individual glassy pieces ranging in size from 0.46 g to 1.95 g) were dissolved, purified for molybdenum, and analyzed for isotopic composition. For each sample, an untraced aliquot was used to measure the Mo isotopic composition, while a second aliquot was traced with a ^{96}Mo spike and used to determine the absolute concentration of Mo in the Trinity debris sample by isotope dilution mass spectrometry. A typical aliquot consisted of ~200 mg of debris containing 120-700 ng of stable Mo along with plutonium, uranium, other long-lived fission and activation products (e.g. ^{137}Cs, ^{152}Eu) as well as stable elements from the surrounding environment. The aliquots were purified for molybdenum using a combination of cation exchange and anion exchange chromatography (details of the separation and analytical methods are included in the supporting information).(20-26) In addition to removing radioactive isotopes and major matrix constituents, it was necessary to effectively remove stable Zr and Ru which could negatively affect the accuracy of Mo isotope ratio measurements due to isobaric interferences from $^{92}Zr^{+}$, $^{94}Zr^{+}$, $^{96}Zr^{+}$, $^{96}Ru^{+}$, and $^{98}Ru^{+}$.

Molybdenum isotope ratios were measured using a Thermo Scientific Neptune Plus multicollector inductively coupled plasma mass spectrometer (MC-ICP-MS) and a static measurement routine where each Mo isotope was collected on a Faraday detector. The ^{92}Mo/^{96}Mo ratio is unperturbed by fission decay and was used as a normalizing ratio in the untraced samples to correct for mass-dependent isotope fractionation during analysis. The ^{92}Mo/^{96}Mo value of 0.87867 is published for the reference standard NIST SRM 3134 (27); molybdenum isotopic variations observed in nature in granites and basalts are small relative to current analytical limits.(25, 27-30) Normalizing the isotopic data using an unperturbed ratio in this way corrects for any source of mass-dependent isotope fractionation, but allows for measurement of mass-independent effects like fission inputs from short-lived Zr.(31) As shown in Table 1 and Figure 2a, the Trinity nuclear debris samples and bracketing NIST 3134 standards had ^{94}Mo/^{96}Mo ratios that were identical within uncertainty.[*] However, significantly elevated

[*] Note that neutron capture reactions in the soil and rock may have occurred during the nuclear detonation, resulting in destruction or formation reactions of stable elements. However, for molybdenum these effects are expected to be small relative to the perturbation from fission ingrowth and no variation is observed between the ^{94}Mo/^{96}Mo ratio measured for the Trinity nuclear debris samples and NIST SRM 3134.

$^{95}Mo/^{96}Mo$ and $^{97}Mo/^{96}Mo$ ratios were detected in the Trinity debris samples (Figure 2b). Traced Mo sample fractions were corrected for isotopic fractionation using a standard-sample bracketing routine and were used to determine the total amount of molybdenum in the trinitite samples, which ranged from 591 to 3570 ng/g of debris (Table 1).

Table 1. Molybdenum isotopic compositions measured for five Trinity nuclear debris samples and concentrations of $^{95}Zr_{Mo}$ and $^{97}Zr_{Mo}$ in Trinity nuclear debris samples calculated from Mo isotopic perturbations. Data was normalized to a $^{92}Mo/^{96}Mo$ ratio of 0.87867. The uncertainties expressed represent expanded (k = 2) values.

Sample	**$^{94}Mo/^{96}Mo$**	**$^{95}Mo/^{96}Mo$**	**$^{97}Mo/^{96}Mo$**	**$^{98}Mo/^{96}Mo$**	**Mo conc. (ng/g debris)**
31-1	0.55109(11)	0.95761(16)	0.58101(11)	1.46318(30)	3570(120)
32-1	0.55113(14)	0.95864(19)	0.58213(11)	1.46429(29)	3140(100)
33-1	0.55111(12)	0.95814(20)	0.58166(13)	1.46381(36)	2788(94)
34-1	0.55110(14)	0.96112(18)	0.58492(13)	1.46706(31)	2053(67)
40-1	0.55109(15)	0.95474(20)	0.57774(18)	1.45953(31)	591(19)
NIST 3134	0.55109(5)	0.95205(10)	0.57470(16)	1.45674(28)	

Sample	**$^{95}Zr_{Mo}$ (atoms/g)**	**$^{97}Zr_{Mo}$ (atoms/g)**	**$^{97}Zr/^{95}Zr$**
31-1	$2.10(10) \times 10^{13}$	$2.38(9) \times 10^{13}$	1.13(8)
32-1	$2.17(11) \times 10^{13}$	$2.46(10) \times 10^{13}$	1.13(8)
33-1	$1.77(10) \times 10^{13}$	$2.03(9) \times 10^{13}$	1.14(8)
34-1	$1.96(8) \times 10^{13}$	$2.21(8) \times 10^{13}$	1.13(6)
40-1	$1.63(19) \times 10^{12}$	$1.81(15) \times 10^{12}$	1.11(16)
average (2σ)			1.13(2)

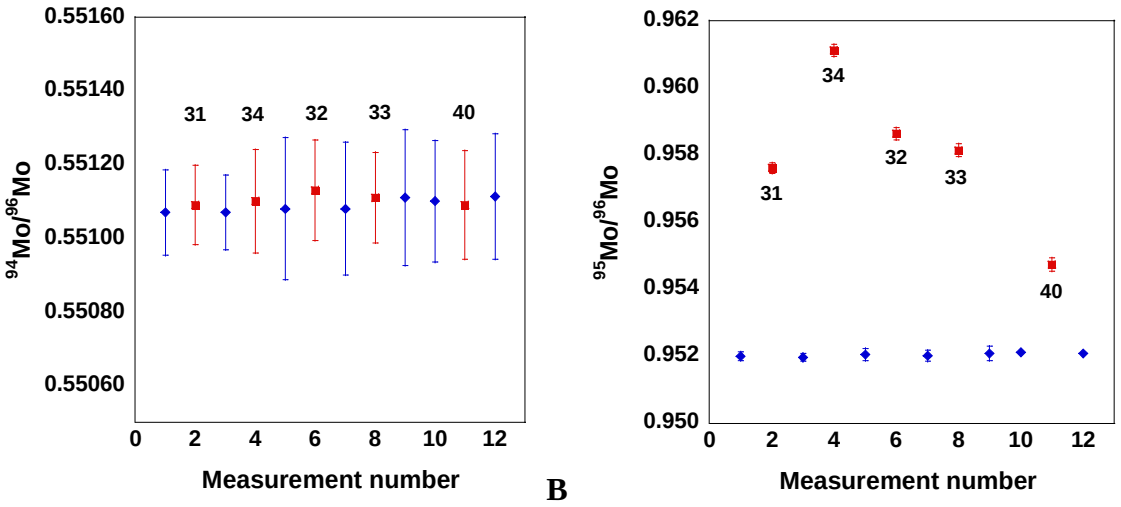

Figure 2. Measurements of $^{94}Mo/^{96}Mo$ (A) and $^{95}Mo/^{96}Mo$ ratios (B), with error bars at 2σ uncertainty. Measurements of Trinity nuclear debris samples are shown with red square while measurements of the reference standard NIST SRM 3134 are shown with blue diamonds. Note the expanded scale of panel A as compared to panel B.

The total number of atoms of ^{95}Zr that would have been present in each debris sample at the time of detonation (denoted $^{95}Zr_{Mo}$) was calculated using the observed $^{95}Mo/^{96}Mo$ ratio, the natural $^{95}Mo/^{96}Mo$ ratio measured for NIST SRM 3134, and the total amount of ^{96}Mo atoms in the sample, as shown in Equation 2. A similar approach was used to calculate the total number of atoms of $^{97}Zr_{Mo}$ in each sample. The average $^{97}Zr/^{95}Zr$ ratio was 1.13(2), consistent with relative cumulative chain yields expected for plutonium fission.(16, 32) This value is higher than the average of 0.928(7) reported by Wieser et al. for the Oklo study, where a uranium thermal fission spectrum was invoked.(18)

$$^{95}Zr_{Mo}(atoms/sample) = \left[\left(\frac{^{95}Mo}{^{96}Mo}\right)_{observed} - \left(\frac{^{95}Mo}{^{96}Mo}\right)_{natural}\right]\left(^{96}Mo(atoms/sample)\right) \qquad \text{(Equation 2)}$$

For comparison, the concentration of ^{137}Cs was determined in each sample via gamma-ray spectroscopy using a high-purity germanium detector (HPGe). Table 2 shows the ^{137}Cs concentrations per gram of sample and associated uncertainties, decay corrected to the test date of 7/16/1945. An aliquot of each Trinity nuclear debris sample was also purified for plutonium by anion exchange chromatography and the concentration and isotopic composition of the plutonium determined by isotope dilution ICP-MS using a ^{242}Pu tracer (NIST SRM 4334G). The decay corrected concentration of ^{239}Pu in each sample is shown in Table 2.

Table 2. Measured concentrations of ^{137}Cs and ^{239}Pu in trinitite, decay corrected to 7/16/1945. The uncertainties expressed represent expanded (k = 2) values.

Sample	**^{137}Cs (atoms/g)**	**^{239}Pu (atoms/g)**
31-1	2.03(14) x 10^{12}	1.72(4) x 10^{15}
32-1	1.07(12) x 10^{12}	1.64(4) x 10^{15}
33-1	1.82(18) x 10^{12}	1.50(3) x 10^{15}
34-1	1.80(15) x 10^{12}	1.64(4) x 10^{15}
40-1	1.70(43) x 10^{11}	1.56(4) x 10^{14}

The number of fissions per gram of sample can be calculated by dividing the measured $^{95}Zr_{Mo}$, $^{97}Zr_{Mo}$, or ^{137}Cs atoms per gram values by the appropriate cumulative fission chain yield for each nuclide. As Trinity was a plutonium fission device, plutonium fission spectrum chain yields are used for ^{95}Zr (4.671%), ^{97}Zr (5.297%) and ^{137}Cs (6.591%).(16) As shown in Equation 1, the device plutonium efficiency is calculated from the measured ^{239}Pu and $^{95}Zr_{Mo}$ or $^{97}Zr_{Mo}$ atoms per sample. For Trinity, the yield can also be calculated by multiplying the known mass of plutonium in Trinity (6 kg) by the efficiency and scaling the resultant total number of device fissions by a term relating fissions to energy release (7.216×10^{-24} kilotons per plutonium fission).(33)

Overall, the refractory zirconium fission products give much more consistent plutonium efficiency values than the volatile ^{137}Cs. The efficiency values determined using ^{137}Cs are a factor of 15 to 20 lower than when zirconium is used, consistent with expected fractionation between ^{137}Cs and ^{239}Pu due to differences in volatility during fireball cooling.(15) The yield values calculated using $^{95}Zr_{Mo}$ and $^{97}Zr_{Mo}$ fission measurements are in excellent agreement with each other and with the official Trinity yield of 21 kt, while the yield calculated using ^{137}Cs as the fission indicator is more than an order of magnitude lower for all five samples (Table 3, Figure 3).

Table 3. Calculated fission efficiency and yield in kilotons of the Trinity device using three different fission indicators following Equation 1. The uncertainties represent expanded (k = 2) values, see Supporting Information for details.

sample	**Calculated Pu Efficiency (%)**			**Calculated Yield (kt)**		
	Using ^{95}Zr	***Using ^{97}Zr***	***Using ^{137}Cs***	***Using ^{95}Zr***	***Using ^{97}Zr***	***Using ^{137}Cs***
31-1	20.5 ± 5%	20.5 ± 5%	1.74 ± 7%	22.3 ± 5%	22.4 ± 5%	1.89 ± 7%
32-1	21.9 ± 6%	21.9 ± 5%	0.97 ± 12%	23.9 ± 6%	23.9 ± 5%	1.06 ± 12%
33-1	20.2 ± 6%	20.4 ± 5%	1.81 ± 10%	22.1 ± 6%	22.2 ± 5%	1.97 ± 10%
34-1	20.4 ± 5%	20.3 ± 4%	1.64 ± 9%	22.2 ± 5%	22.2 ± 4%	1.79 ± 9%
40-1	18.3 ± 12%	18.0 ± 9%	1.63 ± 26%	20.0 ± 9%	19.7 ± 12%	1.78 ± 26%
average (stdev)	20.3(12%)	20.4(12%)	1.56(44%)	22.1(12%)	22.1(12%)	1.70(44%)

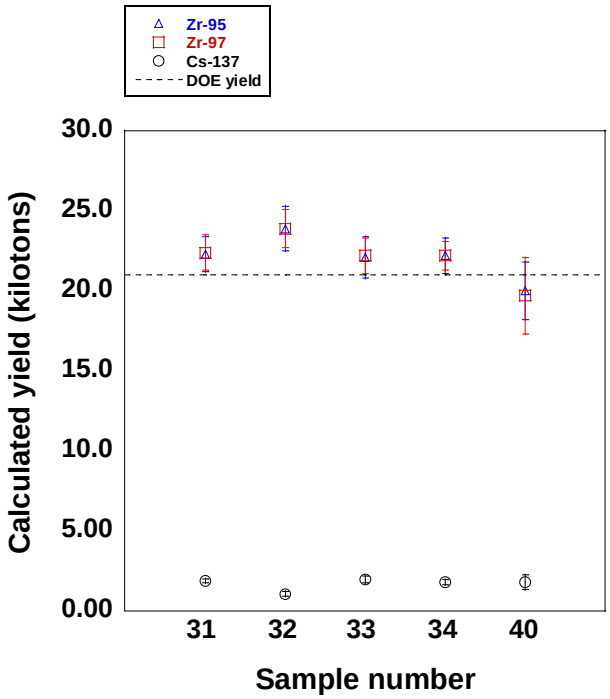

Figure 1. Calculated yield of Trinity using $^{95}Zr_{Mo}$ (blue triangles), $^{97}Zr_{Mo}$ (red squares) and ^{137}Cs (black circles) fission indicators. The dashed gray line shows the official reported yield of Trinity.

Note that the Trinity device did contain a uranium tamper, so some of the ^{239}Pu observed today would arise from neutron capture on ^{238}U to form ^{239}U, which β decays to ^{239}Pu. Additionally, some fissions occurred in the uranium tamper. For these samples, the effects of these complications on the calculated efficiency and yield appear to be small; however, future work will focus on methods to address these factors. The modern yield determination for the Trinity nuclear test, derived from measurements of the extinct $^{95,97}Zr$ fission products is 22.1 ± 2.7 kt.

This work demonstrates that fissions in nuclear bomb debris can be determined quantitatively via high precision mass spectrometric measurements of stable fission product decay daughters. Specifically, decay of the 95 and 97 fission product mass chains to stable molybdenum daughters was quantified via MC-ICP-MS measurements of perturbations in $^{95}Mo/^{96}Mo$ and $^{97}Mo/^{96}Mo$ atom ratios. By combining a measure of fissions in a sample with measurements of residual ^{239}Pu, device fission efficiency can be determined. While ^{137}Cs can be conveniently measured via γ spectroscopy in Trinity nuclear debris samples, it is a poor indicator

of fissions for a nuclear test because it exists predominantly as volatile gaseous species during fireball cooling. Trinity yields calculated from $^{95,97}Zr_{Mo}$ fission measurements are in excellent agreement with the officially reported yield of the Trinity test, and yields calculated using ^{137}Cs measurements are not. This work demonstrates the feasibility of determining plutonium efficiency and device yield in samples many years after the traditionally measured, short-lived fission fragments have decayed below radiometric detection limits. Such technologies could expand the timescale for nuclear inspection and data collection, aiding treaty monitoring and verification endeavors for the foreseeable future.

Materials and methods.

All sample dissolution and purification was performed using Optima grade acids (Fisher Scientific). Savillex PFA jars were used for sample processing. Anion exchange (AG-MP 1M 100-200 mesh) and cation exchange (AG 50WX8 200-400 mesh) resins were obtained from Biorad Laboratories, cleaned prior to use by sequentially washing with 8 M HNO_3, 1.0 M HNO_3/0.5 M HF, 10 M HCl, 6 M HCl, and 1 M HCl, and stored in 0.1 M HCl.

Trinity nuclear debris samples 31-34 were dark gray glassy pieces ranging in size from 0.45 g to 0.63 g obtained from the archives at Los Alamos National Laboratory. Sample 40 was a green glassy piece of 1.95 g obtained from the Mineralogical Research Company of San Jose, CA. Debris samples were dissolved by repeated cycling with HF/HNO_3 or HF/HNO_3/$HClO_4$ and stored as solutions in 3 M HCl or 4 M HNO_3. Molybdenum standard solution NIST SRM 3134 was obtained from NIST (Lot 130418). A ^{96}Mo spike solution was prepared by dissolution of enriched ^{96}Mo metal powder purchased from Trace Sciences International (Lot 134390P). The ^{96}Mo spike was calibrated by reverse isotope dilution against NIST SRM 3134.

^{137}Cs measurements were performed by gamma counting 5 ml of the dissolved Trinity nuclear debris solutions for 1000 minutes on a high-purity germanium detector (Ortec GEM series coaxial HPGe detector). ^{137}Cs concentrations were calculated from the peak area and branching ratio at 661.7 keV and the detector efficiency curve specific to the counting geometry. The total propagated uncertainties on the ^{137}Cs measurements include uncertainties arising from the peak fitting and the calibration source.

Samples were purified for molybdenum using a series of three ion exchange columns. The sample was dissolved in 0.5 M HCl and passed through a cation exchange column (AG 50WX8 200-400 mesh) to remove major matrix constituents. The sample was then evaporated, redissolved in 6 M HCl, and loaded onto an anion exchange column (AG MP 1M 100-200 mesh). The column was washed with 6 M HCl and then Mo was eluted with 1 M HCl. Finally, the sample was passed through an additional cation exchange column in 0.5 M HCl. The three column procedure provided Mo in 60-70% yield and afforded good decontamination against interfering elements, particularly isobars of Zr ($^{92}Zr^+$, $^{94}Zr^+$, $^{96}Zr^+$) and Ru ($^{96}Ru^+$, $^{98}Ru^+$, $^{100}Ru^+$). A typical process blank for the complete purification procedure contained 2 ± 1 ng total Mo.

Molybdenum isotopic measurements were performed using a Thermo Scientific Neptune Plus MC-ICP-MS. A static configuration of eight Faraday cups was used to simultaneously measure $^{90}Zr^+$ (L3), $^{91}Zr^+$ (L2), $^{92}Mo^+$ (L1), $^{94}Mo^+$ (C), $^{95}Mo^+$ (H1), $^{96}Mo^+$ (H2), $^{97}Mo^+$ (H3), and $^{98}Mo^+$ (H4), with 1 x 10^{11} Ohm amplifiers on each cup. Amplifier gain calibration was performed daily using an internally supplied voltage. Isotopes of Mo were included in the instrumental mass calibration by performing peak centering routines at ^{94}Mo and ^{96}Mo using a solution of NIST 3134 before each analysis session. Data were collected in one block of 40 cycles using an 8.134 second/cycle integration time. Each measurement was followed by a rinse protocol involving 40 cycles of 10% HNO_3-0.01 M HF, 40 cycles of 2% HNO_3, and a blank of 40 cycles of 2% HNO_3. The typical sample concentration was 40-100 ppb and the intensity of the $^{98}Mo^+$ signal ranged from 0.5 to 1.1 V measured on a Faraday detector. For all samples, the intensity of the $^{90}Zr^+$ signal was $\leq$ 0.1 mV and no corrections for Zr isobaric interference were performed. NIST 3134 was used for standard-sample bracketing.

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Supporting information for:

Measurements of Extinct Fission Products in Nuclear Bomb Debris: Determination of the Yield of the Trinity Nuclear Test 70 Years Later

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Table S1. Molybdenum purification steps.

Reagent	Volume (ml)
1. Cation exchange column	
Resin: AG 50Wx8 200-400	2
Wash resin: 6 M HCl	10
Condition resin: 0.5 M HCl	10
Load sample/elute: 0.5 M HCl	10
Elute: 0.5 M HCl	10
2. Anion exchange column	
Resin: AG MP 1M 100-200	5
Wash resin: 8 M HNO_3	40
Wash resin: 6 M HCl	50
Wash resin: 1 M HCl	90
Condition resin: 6 M HCl	20
Load sample: 6 M HCl	10
Wash: 6 M HCl	90
Elute: 1 M HCl	60
3. Cation exchange column	
Resin: AG 50Wx8 200-400	2
Wash resin: 6 M HCl	10
Condition resin: 0.5 M HCl	10
Load sample/elute: 0.5 M HCl	10
Elute: 0.5 M HCl	10

Table S2. Molybdenum isotope ratios ratio data for untraced Trinity nuclear debris samples. The uncertainties expressed represent expanded (k = 2) values.

sample	$^{94}Mo/^{96}Mo$	$^{95}Mo/^{96}Mo$	$^{97}Mo/^{96}Mo$	$^{98}Mo/^{96}Mo$
NIST 3134	0.55107(12)	0.95200(14)	0.57465(8)	1.45677(23)
31-1	0.55109(11)	0.95761(16)	0.58101(11)	1.46318(30)
NIST 3134	0.55107(10)	0.95197(11)	0.57462(9)	1.45672(23)
34-1	0.55110(14)	0.96112(18)	0.58492(13)	1.46706(31)
NIST 3134	0.55108(19)	0.95205(18)	0.57464(16)	1.45663(52)
32-2	0.55113(14)	0.95864(19)	0.58213(11)	1.46429(29)
NIST 3134	0.55108(18)	0.95202(17)	0.57466(11)	1.45663(43)
33-1	0.55111(12)	0.95814(20)	0.58166(13)	1.46381(36)
NIST 3134	0.55111(18)	0.95208(22)	0.57472(13)	1.45675(53)
NIST 3134	0.55110(17)	0.95211(19)	0.57479(11)	1.45703(39)
40-1	0.55109(15)	0.95474(20)	0.57774(18)	1.45953(31)
NIST 3134	0.55113(17)	0.95209(22)	0.57483(14)	1.45668(33)
3134 average (stdev)	0.55109(5)	0.95205(10)	0.57470(16)	1.45674(28)

Table S3. Isotope ratio data for the trinitite samples traced with a ^{96}Mo spike. The uncertainties expressed represent expanded (k = 2) values.

sample	$^{92}Mo/^{96}Mo$	$^{94}Mo/^{96}Mo$	$^{95}Mo/^{96}Mo$	$^{97}Mo/^{96}Mo$	$^{98}Mo/^{96}Mo$
NIST 3134	0.8787(5)	0.5510(3)	0.9520(4)	0.5747(3)	1.4568(9)
40-96	0.3359(2)	0.2117(1)	0.3694(1)	0.2264(1)	0.5610(3)
NIST 3134	0.8787(6)	0.5510(4)	0.9520(4)	0.5747(3)	1.4568(7)
NIST 3134	0.8787(7)	0.5510(6)	0.9520(11)	0.5747(7)	1.4568(12)
31-96	0.7501(9)	0.4710(7)	0.8182(10)	0.4972(7)	1.2497(12)
NIST 3134	0.8787(11)	0.5510(7)	0.9520(10)	0.5747(7)	1.4568(13)
NIST 3134	0.8787(6)	0.5510(3)	0.9520(5)	0.5747(3)	1.4568(7)
32-96	0.6126(3)	0.3846(2)	0.6703(3)	0.4082(2)	1.0223(5)
NIST 3134	0.8787(3)	0.5510(3)	0.9520(3)	0.5747(2)	1.4568(7)
34-96	0.5321(2)	0.3343(2)	0.5849(2)	0.3577(2)	0.8905(4)
NIST 3134	0.8787(4)	0.5510(3)	0.9520(3)	0.5747(3)	1.4568(7)
33-96	0.5352(2)	0.3361(1)	0.5867(2)	0.3579(1)	0.8939(3)
NIST 3134	0.8787(3)	0.5510(2)	0.9520(4)	0.5747(3)	1.4568(7)
^{96}Mo spike	0.002082(2)	0.002427(2)	0.010020(4)	0.010493(5)	0.009437(8)

Table S4. Plutonium isotopic measurements, decay corrected to 7/16/1945. The uncertainties expressed represent expanded (k = 2) values.

sample	^{239}Pu (atoms/g debris)	$^{240}Pu/^{239}Pu$	$^{241}Pu/^{239}Pu$
31	$1.72(4) \times 10^{15}$	0.02426(8)	$4.3(8) \times 10^{-4}$
32	$1.64(4) \times 10^{15}$	0.02508(12)	$5.2(6) \times 10^{-4}$
33	$1.50(3) \times 10^{15}$	0.02432(12)	$4.6(8) \times 10^{-4}$
34	$1.64(4) \times 10^{15}$	0.02446(14)	$4.8(8) \times 10^{-4}$
40	$1.56(4) \times 10^{14}$	0.02476(20)	$4.7(8) \times 10^{-4}$

Calculations

Device efficiency was calculated according to the following equation, using measured values for ^{239}Pu and $^{95}Zr_{Mo}$ and $^{97}Zr_{Mo}$.

$$Efficiency = \frac{F_{Total}}{Pu_{Ingoing}} = \frac{f_{Sample}}{f_{Sample} + Pu_{Sample}}$$

F_{Total} = total number of fissions in the device
$Pu_{Ingoing}$ = total number of plutonium atoms in the device
f_{sample} = number of fissions measured in the debris sample ($^{95}Zr_{Mo}$ or $^{97}Zr_{Mo}$ atoms/g debris)
Pu_{sample} = number of plutonium atoms measured in the debris sample (^{239}Pu atoms/g debris)

The uncertainty in the efficiency was calculated as follows, using the expanded (k = 2) measurement uncertainties for ^{239}Pu and $^{95}Zr_{Mo}$ and $^{97}Zr_{Mo}$.

$$\frac{\sigma_{Efficiency}}{Efficiency} = \sqrt{\left(\frac{\sigma_{Fissions}}{Fissions}\right)^2 + \left(\frac{\sqrt{{\sigma_{Pu}}^2 + {\sigma_{Fissions}}^2}}{Pu + Fissions}\right)^2}$$